

Amendments to the Claims

This listing of claims will replace all prior versions, and listings, of claims in the application.

Listing of Claims

1. (Currently amended) A continuous staged countercurrent process that obtains improved oxygen utilization and desirable product quality for the catalytic oxidation of at least one benzene disubstituted with oxidizable substituents selected from the class consisting of alkyl, hydroxyalkyl, aldehyde, carboxy groups and mixtures thereof, to its corresponding dicarboxylic acid in a solvent system, comprising the steps of:

- (a) introducing into a first oxidation stage a feed mixture comprising at least a portion of the total amount of each of (i) the solvent comprising an organic acid, (ii) at least one catalytic metal selected from the class consisting of manganese, cobalt, nickel, zirconium, hafnium, cerium and mixtures thereof, and (iii) bromine at a mole ratio based on total catalytic metals within the range of from about 1:20 to about 5:1, and from about 7 to about 60 weight percent of the total amount of the at least one disubstituted benzene introduced into the first oxidation stage and into a second oxidation stage;
- (b) partially oxidizing the at least one disubstituted benzene in the first oxidation stage with the presence of a molecular oxygen-containing gas initially containing from about 3 to about 20 volume percent of molecular oxygen, at a temperature in the range of from about 250°F to about 401°F and with the relative amounts of disubstituted benzene, catalytic metal, solvent and bromine and with the temperature being such that from about 25 to about 99.95 weight percent of the disubstituted benzene fed to the first oxidation stage is oxidized to form a gas mixture comprising unreacted molecular oxygen and vaporized solvent and a first product mixture comprising dicarboxylic acid product, partially oxidized disubstituted benzene and unreacted disubstituted benzene and solvent, and at a pressure in the range of from about 130 to about 215 psig to maintain the disubstituted benzene, partially oxidized disubstituted benzene, dicarboxylic acid product and solvent as a liquid or solid-liquid slurry, and such that the concentration of residual molecular oxygen in the remaining gas mixture

is from about 0.3 to about 2 volume percent thereby obtaining improved oxygen utilization;

(c) recovering the resulting first product mixture from the first oxidation stage and feeding at least a portion of the recovered first product mixture to the second oxidation stage;

(d) removing the remaining gas mixture from the first oxidation stage and partially condensing the gas mixture so as to remove therefrom condensable solvent and at least a portion of the condensed solvent is introduced into the first oxidation stage, the second oxidation stage, or both thereof;

(e) feeding to the second oxidation stage a gas containing molecular oxygen and the portion not fed into the first oxidation stage, if any, of the aforesaid total amounts introduced in steps (a) and (e) of disubstituted benzene, catalytic metal, solvent and bromine;

(f) oxidizing in the second oxidation stage the partially oxidized disubstituted benzene and unreacted disubstituted benzene fed to the second oxidation stage with a molecular oxygen-containing gas containing from about 15 to about 50 volume percent of molecular oxygen at a temperature within the range of from about 347°F to about 421°F, and with the relative amounts of disubstituted benzene, partially oxidized disubstituted benzene, catalytic metal, solvent and bromine and with the temperature being such that from about 96 to about 100 weight percent of the disubstituted benzene and partially oxidized disubstituted benzene are oxidized to form a gas mixture comprising unreacted molecular oxygen and vaporized solvent and a second product mixture comprising the dicarboxylic acid product and solvent, and at a pressure in the range of from about 170 to about 235 psig to maintain the dicarboxylic acid product, partially oxidized disubstituted benzene and unreacted disubstituted benzene as a liquid or solid-liquid slurry and such that the concentration of residual molecular oxygen in the remaining gas mixture is in the range of from about 3 to about 15 volume percent;

(g) recovering from the second oxidation stage the second product mixture comprising the dicarboxylic acid product having an optical density at 340 nm of less than about 1.0; and

- (h) withdrawing from the second oxidation stage and recycling to the first oxidation stage the residual molecular oxygen containing gas.
2. (Previously amended) The process of Claim 1 wherein the disubstituted benzene is para-disubstituted benzene and the corresponding dicarboxylic acid is terephthalic acid.
3. (Original) The process of Claim 2 wherein the substituents in the para-disubstituted benzene are alkyl groups having from one to four carbon atoms.
4. (Canceled)
5. (Previously amended) The process of Claim 1 wherein the solvent comprises acetic acid.
6. (Original) The process of Claim 1 wherein the catalytic metals are cobalt and manganese.
7. (Original) The process of Claim 1 wherein the atom ratio of manganese to cobalt in the reaction mixture in the first oxidation stage is in the range of from about 1:100 to about 100:1.
8. (Original) The process of Claim 1 wherein the reaction mixture in the first oxidation stage mixture comprises a mole ratio of bromine-to-total catalytic metals therein of from about 1:5 to about 2:1.
9. (Original) The process of Claim 1 wherein the temperature in the first oxidation stage is maintained within the range of from about 277°F to about 351°F.
10. (Original) The process of Claim 1 wherein the molecular oxygen-containing gas introduced in the first oxidation stage contains from about 3 to about 11 volume percent of molecular oxygen.
11. (Original) The process of Claim 1 wherein the concentration of residual molecular oxygen in the gas removed from the first oxidation stage is less than about 1 volume percent.
12. (Previously amended) The process of Claim 1 wherein the degree of conversion of the disubstituted benzene to partially oxidized disubstituted benzene and dicarboxylic acid thereof in the first oxidation stage is within the range of from about 60 to about 99.95 weight percent.

13. (Original) The process of Claim 1 wherein the molecular oxygen-containing gas introduced into the second oxidation stage contains from about 20 to about 25 volume percent of molecular oxygen.
14. (Original) The process of Claim 1 wherein the concentration of residual molecular oxygen in the gas removed from the second oxidation stage is from about 3 to about 11 volume percent.
15. (Original) The process of Claim 14 wherein the concentration of residual molecular oxygen in the gas removed from the second oxidation stage is from about 3 to about 8 volume percent.
16. (Previously amended) The process of Claim 1 wherein the degree of conversion of the disubstituted benzene and partially oxidized disubstituted benzene to the dicarboxylic acid thereof in the second oxidation stage is within the range of from about 97 to about 100 weight percent.
17. (Previously amended) The process of Claim 6 wherein from about 20 to about 100 weight percent of the aforesaid total amount of manganese added in steps (a) and (d) is added in the first oxidation stage.
18. (Previously amended) The process of Claim 6 wherein from about 20 to about 100 weight percent of the aforesaid total amount of cobalt added is added in steps (a) and (d) in the first oxidation stage.
19. (Previously amended) The process of Claim 1 wherein from about 20 to about 100 weight percent of the total aforesaid amount of bromine added in steps (a) and (d) is added in the first oxidation stage.
20. (Previously amended) The process of Claim 1 wherein from about 15 to about 35 weight percent of the aforesaid total amount of disubstituted benzene added in steps (a) and (d) is added in the first oxidation stage.
21. (Previously amended) The process of Claim 1 wherein from about 10 to about 100 weight percent of the total aforesaid amount of solvent added in steps (a) and (d) is added in the first oxidation stage.
22. (Original) The process of Claim 1 wherein the temperature in the first oxidation stage is at least 5.5°F lower than the temperature in the second oxidation stage.
23. (Canceled)

24. (Canceled)
25. (Canceled)
26. (Previously amended) The process of Claim 1 wherein the condensed solvent is introduced into the second oxidation stage.
27. (Original) The process of Claim 1 wherein the second product mixture recovered from the second oxidation stage is subjected to oxidation conditions in a third oxidation stage to thereby oxidize oxidizable impurities therein.
28. (Previously amended) The process of Claim 27 wherein the oxidized impurities are separated from the dicarboxylic acid product to thereby produce a purified dicarboxylic acid product.
29. (Canceled)
30. (Canceled)